

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE****Patentee:** Shen et al.**Assignee:** Atwood Mobile Products, Inc.**U.S. Patent No.:** 5,573,648 **Date Issued:** November 12, 1996**Reissue Application No.:** 10/621,999 **Date Filed:** July 17, 2003**Application No.:** 381,718 **Date Filed:** January 31, 1995**Title:** GAS SENSOR BASED ON PROTONIC CONDUCTIVE MEMBRANES

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Mail Stop Reissue - Amendment  
Commissioner for Patents  
P.O. Box 1450  
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**SUPPLEMENTAL DECLARATION OF  
COMMERCIAL SUCCESS UNDER 37 C.F.R. § 1.132**

Dear Sir:

I, Yousheng Shen, declare as follows:

1. I am the former Electrochemical Research Laboratory Manager for Atwood Mobile Products ("Atwood"). I was employed by Atwood from November 1994 to November 2007. My daily duties included development of new electrochemical-related products.
2. I am currently employed at Delta International as Engineering Director. I am being paid as a consultant by Atwood for assisting in the preparation and presentation of this Supplemental Declaration.

3. I received a doctorate degree in Material Science and Engineering from the University of Utah in 1994. My research as a doctoral candidate was directed towards ionic and electronic mixed conductors.
4. I understand that United States Patent No. 5,573,648 (the '648 Patent) and related patent, United States Patent No. 5,650,054 (the '054 Patent), both are the subject of reissue proceedings in the United States Patent and Trademark Office. The reissue application of the '648 Patent is application no. 10/621,999 (the '999 Reissue Application). The reissue application of the '054 Patent is application no. 621,637 (the '637 Reissue Application). I am a named inventor on the '648 and '054 Patents.
5. The '648 and '054 Patents are directed to improved gas sensors. The '648 Patent is aptly titled "Gas Sensor Based on Protonic Conductive Membranes." Described in the specification of the '648 Patent are electrochemical gas sensors having a protonic conductive solid electrolyte membrane.
6. Claim 1 of the '999 Reissue Application (as currently amended) reads:
  1. An electrochemical gas sensor for quantitative measurement of a gas in an ambient atmosphere at room temperature comprising:
    - a porous mixed ionic-electronic conductive sensing electrode having both an electronic conducting material and an ionic conducting material;
    - a porous mixed ionic-electronic conductive counter electrode having both an electronic conducting material and an ionic conducting material;
    - a first protonic conductive solid electrolyte membrane in between and in contact with the sensing and counter electrodes, and having a thickness in the range of approximately 0.1 mm to 1 mm;
    - the sensing electrode being capable of reacting with the gas to produce a change in an electrical characteristic between the sensing electrode and the counter electrode;
    - means for electrical measurement electrically connected to said sensing and counter electrodes;
    - said sensing and counter electrodes each having a diameter in the range of approximately 1 mm to 15 mm, and being electrically connected to said electrical measurement means;

whereby said electrical measurement means is capable of detecting changes in said electrical characteristic in a positive ambient atmosphere concentration of said gas at room temperature.

New claim 97 of the '999 Reissue Application reads:

97. A two-electrode electrochemical gas sensor for quantitative measurement of a carbon monoxide gas in an ambient atmosphere at room temperature comprising:

a porous mixed ionic-electronic conductive sensing electrode having both an electronic conducting material and an ionic conducting material, the sensing electrode including platinum, carbon and a copolymer having a tetrafluorethylene backbone with a side chain of perfluorinated monomers containing at least one of a sulfonic acid group;

a porous mixed ionic-electronic conductive counter electrode having both an electronic conducting material and an ionic conducting material, the counter electrode including platinum, carbon and a copolymer having a tetrafluorethylene backbone with a side chain of perfluorinated monomers containing at least one of a sulfonic acid group;

a first protonic conductive solid electrolyte membrane in between and in contact with the sensing and counter electrodes, and having a thickness in the range of approximately 0.1 mm to 1 mm, the protonic conductive solid electrolyte membrane being substantially comprised of a solid, perfluorinated, ion-exchange polymer and being approximately 0.17 mm thick;

the sensing electrode being capable of reacting with the carbon monoxide gas to produce a change in an electrical characteristic between the sensing electrode and the counter electrode, the sensing electrode and the counter electrode being the only two electrodes in contact with the first protonic conductive electrolyte membrane and the sensing electrode and the counter electrode being on opposite sides of the first protonic conductive electrolyte membrane;

means for electrical measurement electrically connected to said sensing and counter electrodes;

said sensing and counter electrodes each having a diameter in the range of approximately 1 mm to 15 mm, and being electrically connected to said electrical measurement means, the sensing electrode being 15 mm in diameter and the counter electrode being approximately 15 mm in diameter;

whereby said electrical measurement means is capable of detecting changes in said electrical characteristic in a positive ambient atmosphere concentration of said gas at room temperature.

7. I examined examples of the commercial embodiments of the carbon monoxide (CO) sensors sold by the licensee (the "CO Sensors") at the beginning of the licensing period.

- a) As a general overview, the CO Sensors detect CO, at room temperature under ambient atmosphere conditions, by detecting a change in an electrical current.
- b) The CO Sensors have a sensing electrode and a counter electrode on opposite sides of a solid electrolyte membrane.
- c) The sensing and counter electrodes are the only two electrodes in contact with this membrane.
- d) The sensing electrode is made of platinum, carbon and a copolymer having a tetrafluorethylene backbone with a side chain of perfluorinated monomers containing at least one of a sulfonic acid group (hereinafter "Nafion").
- e) The platinum and the carbon in the sensing electrode are electronic conducting materials.
- f) The Nafion in the sensing electrode is an ionic/protonic conducting material.
- g) The sensing electrode is 15 mm in diameter.
- h) The counter electrode is made of platinum, carbon and Nafion.
- i) The platinum and the carbon in the counter electrode are electronic conducting materials.
- j) The Nafion in the counter electrode is an ionic/protonic conducting material.
- k) The counter electrode is 15 mm in diameter.
- l) The solid electrolyte membrane is a protonic conductive solid electrolyte membrane made of Nafion.
- m) The solid electrolyte membrane is 0.17 mm thick.
- n) When the sensing electrode comes into contact with CO gas, ionized hydrogen atoms (i.e., protons) and electrons are produced. Both the protons and the electrons travel from the sensing electrode where they are produced to the counter electrode.
- o) The protons travel via the solid electrolyte membrane to the counter electrode.
- p) The electrons travel via an external electrical circuit to the counter electrode, thereby allowing the current generated by the sensing electrode to be measured.

- q) A water vapor reservoir supplies water vapor to the sensing electrode, counter electrode and solid electrolyte membrane.<sup>1</sup>

8. I have determined that the CO Sensors are covered by the instant claims of the '999 Reissue Application.

a) The CO Sensors are electrochemical gas sensors for measurement (including quantitative measurement) of a gas in an ambient atmosphere at room temperature (as recited in the preamble of claims 1 and 97).

1) Specifically, the CO Sensors detect carbon monoxide (see, *e.g.*, the further limitations of claims 8 and 97).

2) Further, in the CO Sensors, the sensing electrode and the counter electrode are the only two electrodes in contact with the first protonic conductive electrolyte membrane (see, *e.g.*, the further limitations of claims 79 and 97).

b) The CO Sensors have a porous mixed ionic-electronic conductive sensing electrode having both an electronic conducting material and an ionic conducting material (as recited in claims 1 and 97).

1) The sensing electrode includes carbon, a noble metal or a metal oxide (see, *e.g.*, the further limitations of claims 2, 3 and 4).

2) Specifically, the sensing electrode includes platinum, carbon and a copolymer having a tetrafluorethylene backbone with a side chain of perfluorinated monomers containing a sulfonic acid group (see, *e.g.*, the further limitations of claim 97).

3) More specifically, the electronic and ionic conducting materials of the sensing electrode are a proton-electron mixed conductive material having 10-50 wt% of a

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<sup>1</sup> The CO Sensor includes a water vapor reservoir. The claims in this Reissue Application do not include any limitations directed to an exposure of the sensor components to water vapor. As such, this Declaration does not rely on the contribution of a supply of water vapor to the commercial success of the CO Sensor. The inclusion of a water vapor reservoir in the commercial embodiment is only one feature among several features that contribute to the commercial success of the CO Sensor. As the Examiner is most likely aware, Applicants' currently co-pending Merged Reissue/Reexamination applications serial nos. 10/621,637 and 90/006,209 contain claims that recite "water vapor."

- proton conductor material and 50-90 wt% of first and second electrical conductor materials (see, *e.g.*, the further limitations of claim 14).
- 4) The proton conductor material for the sensing electrodes is a copolymer having a tetrafluorethylene backbone with a side chain of perfluorinated monomers containing a sulfonic acid group (see, *e.g.*, the further limitations of claims 15 and 97).
  - 5) Even more specifically, one of the first and second electrical conductor materials for the sensing electrode is 50-99 wt% of carbon black, and the other of the first and second electrical conductor materials for the sensing electrode is 1-50 wt% of platinum (see, *e.g.*, the further limitations of claim 16).
- c) The CO Sensors have a porous mixed ionic-electronic conductive counter electrode having both an electronic conducting material and an ionic conducting material (as recited in claim 1).
- 1) The counter electrode includes carbon, a noble metal and a metal oxide (see, *e.g.*, the further limitations of claims 2, 3 and 4).
  - 2) Specifically, the counter electrode includes platinum, carbon and a copolymer having a tetrafluorethylene backbone with a side chain of perfluorinated monomers containing a sulfonic acid group (see, *e.g.*, the further limitations of claim 97).
  - 3) More specifically, the electronic and ionic conducting materials of the counter electrode are a proton-electron mixed conductive material having 10-50 wt% of a proton conductor material and 50-90 wt% of a first and a second electrical conductor materials (see, *e.g.*, the further limitations of claim 14).
  - 4) The proton conductor material for the counter electrode is a copolymer having a tetrafluorethylene backbone with a side chain of perfluorinated monomers containing sulfonic acid group (see, *e.g.*, the further limitations of claims 15 and 97).
- d) The CO Sensors have a first protonic conductive solid electrolyte membrane in between and in contact with the sensing and counter electrodes, and having a thickness in the range of approximately 0.1 mm to 1 mm (as recited in claims 1 and 97).

- 1) Specifically, the protonic conductive electrolyte membrane is substantially comprised of a solid, perfluorinated, ion-exchange polymer (see, *e.g.*, the further limitations of claims 5 and 97).
  - 2) Specifically, the protonic conductive electrolyte membrane has a thickness of approximately 0.17 mm (see, *e.g.*, the further limitations of claims 13 and 97).
  - 3) Further, the sensing electrode and the counter electrode are on opposite sides of the first protonic conductive electrolyte membrane (see, *e.g.*, the further limitations of claims 82 and 97).
- e) The sensing electrode of the CO Sensor is capable of reacting with the gas to produce a change in an electrical characteristic between the sensing electrode and the counter electrode (as recited in claims 1 and 97).
- 1) Specifically, the sensing electrode is capable of reacting with carbon monoxide (CO) (see, *e.g.*, the further limitations of claims 8 and 97).
  - 2) Further, the sensing electrode reacts with the gas to produce a change in an electrical characteristic between the sensing electrode and the counter electrode in the absence of an applied voltage to the sensing electrode (see, *e.g.*, the further limitations of claim 80).
- f) The CO Sensor includes means for electrical measurement that are electrically connected to the sensing and counter electrodes (as recited in claims 1 and 97).
- g) The sensing and counter electrodes each having a diameter in the range of approximately 1 mm to 15 mm, and are electrically connected to the electrical measurement means (as recited in claims 1 and 97).
- 1) Specifically, the sensing and counter electrodes have a diameter of approximately 15 mm (see, *e.g.*, the further limitations of claim 97).
  - 2) The electrical measurement means is capable of detecting changes in the electrical characteristic in a positive ambient atmosphere concentration of the gas at room temperature (see, *e.g.*, the further limitations of claims 1 and 97).
  - 3) In a positive ambient concentration of the gas at room temperature, the electrical measurement means is capable of detecting changes in the electrical characteristic in the absence of any biasing voltage (see, *e.g.*, the further limitations of claim 92).

9. As set forth in the specification of the '999 Reissue Application, the invention represents significant improvements over prior art carbon monoxide and toxic gas sensors. Commercial embodiments of the claimed invention, manufactured by Atwood's licensee under the patent, i.e. the CO Sensors, exhibit many commercially significant improvements over prior art carbon monoxide sensors:
- i) the CO Sensors operate reliably at room temperature;
  - ii) the CO Sensors do not need recalibration during their lifetime (typically 5 years or more);
  - iii) the CO Sensors, themselves, do not consume any power;
  - iv) the CO Sensors have an improved CO detection accuracy; and
  - v) the CO Sensors are cheaper to manufacture.
10. The commercial advantages listed in Section 9 are related to and made possible by the following technical performance properties and claimed features:
- i) the CO Sensors operate reliably and rapidly at room temperature, in part, because of their strong current signal output in response to detection of CO, which is due to the combination of:
    - a) the sensing electrode, the counter electrode and the solid electrolyte membrane all including a protonic conductor, and thereby having a minimal ionic/protonic ( $H^+$ ) resistance between the electrodes; and further
    - b) the use of mixed ionic-electronic conductive sensing and counter electrodes, thereby providing a high surface area for three-phase interface contact to occur; and further
    - c) the short ion path (i.e., the thickness of the solid electrolyte membrane being from between 0.1 mm and 1.0 mm); and further
    - d) the sensing and the counter electrodes having diameters in the range of from approximately 1 mm to 15 mm.
  - ii) the CO Sensors do not need recalibration during their lifetime (typically 5 years or more), because their baseline calibration of 0 ppm of CO (i.e., clean air) does not shift



regardless of high temperatures or contamination of the electrolyte and because the continuity in transport of electrical charges avoid polarization effects at the electrodes, which are due to the combination of:

- (a) operating at room temperature; and further
  - (b) having a solid electrolyte membrane; and further
  - (c) the use of mixed ionic-electronic conductive sensing and counter electrodes, thereby providing a high surface area for three-phase interface contact to occur.
- iii) the CO Sensors, themselves, do not consume any power because they do not require a reference electrode and amplifier, thereby eliminating the need for applied DC power to drive the detection current, which is due to the combination of:
- a) the sensing electrode, the counter electrode and the solid electrolyte membrane all including a protonic conductor, and thereby having a minimal ionic/protonic ( $H^+$ ) resistance between the electrodes; and further
  - b) the use of mixed ionic-electronic conductive sensing and counter electrodes, thereby providing a high surface area for three-phase interface contact to occur; and further
  - c) the short ion path (i.e., the thickness of the solid electrolyte membrane being from between 0.1 mm and 1.0 mm); and further
  - d) the sensing and the counter electrodes having diameters in the range of from approximately 1 mm to 15 mm.
- v) the CO Sensors have an improved CO detection accuracy because of their strong current signal output in response to detection of CO and because there is no significant diffusion of contaminating hydrogen molecules, which are due to the combination of:
- a) the sensing electrode, the counter electrode and the solid electrolyte membrane all including a protonic conductor, and thereby having a minimal ionic/protonic ( $H^+$ ) resistance between the electrodes; and further
  - b) the use of mixed ionic-electronic conductive sensing and counter electrodes, thereby providing a high surface area for three-phase interface contact to occur; and further

- c) the short ion path (i.e., the thickness of the solid electrolyte membrane being from between 0.1 mm and 1.0 mm); and further
  - d) the sensing and the counter electrodes having diameters in the range of from approximately 1 mm to 15 mm; and further
  - e) having a solid electrolyte membrane.
- vi) the CO Sensors are cheaper to manufacture, for example, by using an automatic insertion production line at 30 products per minute, due to the combination of:
- a) not needing a reference electrode, an amplifier, a DC power source for the CO Sensor, itself; and further
  - b) the simplicity of using a solid electrolyte membrane.

Each of the above-listed technical performance properties, which make the licensed CO Sensor commercially successful, result from the inventive geometry and selection of materials claimed in the '999 Reissue Application. As presented above, these interrelated technical performance properties are directly attributable to limitations in the claimed subject matter of the '999 Reissue Application.

11. From my communications with Atwood executives, (and/or that of its former corporate parent, Dura Automotive Industries, Inc.), I am informed and believe that the '648 Patent and the '054 Patent are the subject of a royalty-bearing license agreement ("License") entered into in June 1998. The License is an arms-length agreement between separate corporate entities. I am further informed and believe that Atwood has received royalty payments and royalty reports pursuant to the License that set forth the number of CO Sensors sold by the licensee under the License.
12. I am informed and believe that a declaration regarding the commercial success of the invention covered by the claims of the '648 Patent was filed previously in the '648 re-examination proceeding; that the prior declaration states that patented CO Sensors were first sold under the License in November 1998; that according to such royalty reports, 1,991,639 patented CO Sensors were sold under the License in 1999, the first full year

under the License; that according to such royalty reports, 2,717,913 patented CO Sensors were sold under the License in 2001, which represented over a 26.7% increase in sales over the three-year period from 1999 to 2001.

13. I am informed and believe that Atwood has continued to receive substantial royalty payments under the License for sales of patented CO Sensors. I am informed and believe that royalty payments under the License since 2001 have amounted to:

2002	\$536,845
2003	\$677,387
2004	\$889,961
2005	\$815,620
2006	\$989,744
2007	\$674,741

In other words, royalty payments received after 2002 have exceeded four million five hundred thousand dollars (\$4,500,000).

14. I believe that the market share of the CO Sensors in North America today is about 70% of the total market share of residential carbon monoxide sensors.
15. The commercial success of the patented CO Sensors sold pursuant to the License is directly attributable to the advantageous features set forth above in this declaration, which are directly attributable to the features claimed in the '999 Reissue Application.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true and further that these statements were made with knowledge that willful false statements, and the like, so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the above-referenced patent.

Date: September 19, 2008



Dr. Yousheng Shen